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(54) **Title:** ROD–SHAPED PELLETS

(57) **Abstract**

Rod–shaped pellets containing a thermoplastic polymer and glass fibres, with the glass fibres extending in the longitudinal direction of the pellet and having the length of the pellet, wherein the pellets have a core and a sleeve, with the core containing a mixture of glass fibres and polypropylene fibres, with the polypropylene of the fibres being a homopolymer or a random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1–olefins with 4–10 C–atoms and dienes with 4–10 C–atoms, the polypropylene of the fibres having a melt index (230 °C/2.16 kg) of 5 – 500 dg/min. and a density of at least 900 kg/m^3, with at least 50 wt.% of the sleeve consisting of a polypropylene homopolymer, random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1–olefins with 4–10 C–atoms and dienes with 4–10 C–atoms or a block copolymer of propylene and at most 27 wt.% of one or more olefins from the group ethylene and butene and at most 8 wt.% of one or more olefins from the group 1–olefins with 3–10 C–atoms and dienes with 4–10 C–atoms and the propylene contained in the sleeve having a melt index of 1 – 200 dg/min. (230 °C/2.16 kg) and a density of at least 900 kg/m^3.

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ROD-SHAPED PELLETS

The invention relates to rod-shaped pellets containing a thermoplastic polymer and glass fibres, with the glass fibres extending in the longitudinal direction of the pellets and having the length of the pellets.

Pellets of this type are known from EP-A-170245.

The above-mentioned patent application describes a granulate containing a thermoplastic polymer with a very low viscosity. The granulate has been made by pulling a bundle of glass fibres through a melt of the polymer, so that the bundle of glass fibres is impregnated with the melt, then cooling the resulting strand of glass fibres and thermoplastic polymer and reducing it to a granulate.

Because of the low viscosity of the polymer, the polymer melt penetrates the bundle of glass fibres during the production of the granulate, so that every or virtually every single glass fibre is surrounded by the polymer. As the glass fibres are surrounded by the polymer, the strand has a very high strength and shaped articles made from the granulate have very good mechanical properties. This is because during the shaping of the granulate into shaped articles the glass fibres are dispersed very well and glass fibre breakage occurs to only a small extent.

However, the manufacture of the pellets from EP-A-170245 is very laborious, mainly because the glass fibre bundles can be pulled through the melt at only a low speed.

The object of the invention is to provide rod-shaped pellets which are easier to manufacture and which can nevertheless yield shaped articles with good mechanical properties.

Surprisingly, this object is achieved in that pellets are provided which have a core and a sleeve, with the core containing a mixture of glass fibres and polypropylene fibres, with the polypropylene of the fibres being a homopolymer or a random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1-olefins with 4-10 C-atoms and dienes with 4-10 C-atoms, the polypropylene of the fibres having a melt index (230°C/2.16 kg) of 5 - 500 dg/min. and a density of at least 900 kg/m^3, with at least 50 wt.% of the sleeve consisting of a polypropylene homopolymer, random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1-olefins with 4-10 C-atoms and dienes with 4-10 C-atoms or a block copolymer of propylene and at most 27 wt.% of one or more olefins from the group ethylene and butene and at most 8 wt.% of one or more olefins from the group 1-olefins with 3-10 C-atoms and dienes with 4-10 C-atoms and the propylene contained in the sleeve having a melt index of 1 - 200 dg/min. (230° C/2.16 kg) and a density of at least 900 kg/m^3.

A granulate of this type can easily be manufactured, for example by covering a bundle containing the glass fibres and the polypropylene fibres with the aid of a cable-covering unit, cooling the resulting strand and reducing it to a granulate. It has been found that very high production rates can be achieved in this way, which means that the pellets can be manufactured on an industrial scale.

It has moreover been found that if the granulate is processed into a shaped article by melting the pellets in an extruder and extruding them into an object, placing the object in an opened mould in molten condition and closing the mould to compress the shaped article, the shaped article thus obtained has very good mechanical properties. In particular, the shaped object

has a very high impact resistance. This means that the pellets are very suitable for the manufacture of structural mouldings.

The fact that the good mechanical properties are achieved with the aid of the pellets according to the invention is highly surprising, the more so as the glass fibres in the pellets are not at all surrounded by polymer and EP-A-170245 specifically teaches that this is necessary.

From EP-A-226420 a flexible, rod-shaped composite is known which comprises a core of reinforcing fibres and polymer fibres and a thin sleeve of a thermoplastic polymer, which composite can be reduced to pellets. Carbon fibres are recommended as reinforcing fibres. However, the pellets according to the invention are not described in EP-A-226420, nor can it be derived from this document that the pellets according to the invention are easy to produce and that the shaped articles moulded from the pellets can have the good mechanical properties.

The value of the melt index of the polypropylene of the fibres is preferably higher than that of the polymer composition of the sleeve. This results in very homogeneous mouldings with even better mechanical properties.

For the glass fibres in the pellets the usual fibres can be used, which usually have a thickness of between 8 and 25 mμ. The glass fibres preferably have a thickness of between 15 and 20 mμ.

The polypropylene fibres can have the usual thickness for polypropylene fibres of for example 5 - 35 mμ, preferably 10-30 mμ. The polypropylene of the polypropylene fibres preferably has a melt index of 10 - 300 dg/min (230°C/2.16 kg), even more preferably 20 - 100 dg/min. Besides polypropylene, the fibres may contain the usual additives, such as stabilizers, processing aids, nucleating agents and pigments. The

polypropylene used is preferably a homopolymer.

The glass fibres and the polypropylene fibres may be present in separate bundles of for example 400 - 4000 fibres each in the pellets according to the invention. However, the fibres are preferably distributed as homogeneously as possible, so that the glass fibres are present as single fibres and/or in groups of fibres with at most 300 fibres, preferably at most 100 fibres, still more preferably at most 50 fibres. Even more preferably, the fibres are distributed in such a way that at most 50% of the fibres are entirely surrounded by glass fibres. A glass fibre is entirely surrounded by glass fibres, if all fibres that are immediate neighbours of the glass fibre concerned are themselves glass fibres.

The core of the pellets according to the invention preferably contains 50-95 wt.% glass fibres and 50-5 wt.% polypropylene fibres. More preferably, the core contains 60-85 wt.% glass fibres and 40-15 wt.% polypropylene fibres.

The sleeve preferably contains 75-100 wt.% polypropylene, more preferably the sleeve contains 85-100 wt.% polypropylene.

The sleeve preferably contains a polypropylene homopolymer or block copolymer containing a block containing 94-100 wt.% propylene monomer units and 6-0 wt.% ethylene monomer units and a block containing 20 - 80 wt.% ethylene monomer units and 80 - 20 propylene monomer units, with the block copolymer containing a total of 1-25 wt.% ethylene.

It is also possible for the sleeve to contain, besides polypropylene, an elastomer, for example a copolymer of ethylene with a 1-olefin having 3-10 C-atoms, or for example a polymer of ethylene, propylene and one or more dienes, for example EPDM.

The pellets preferably contain 10-70 wt.% glass fibres and a total of 90-30 wt.% polypropylene

and elastomer, more preferably the pellets contain 30-50 wt.% glass fibres and a total of 50-70 wt.% polypropylene and elastomer.

In a special embodiment the pellets contain 0.01-4.0 wt.% carbon black. The pellets preferably contain 0.1-1.0 wt.% carbon black. Preferably, both the sleeve and the polypropylene fibres from the core contain the carbon black. Pellets that contain glass fibres need to contain carbon black for a number of applications because for the shaped articles in question a black colour is required. A disadvantage of this, however, is that the mechanical properties of the shaped articles have a low value compared with shaped articles made from granulate with glass fibres that do not contain the carbon black. However, it is highly surprising that this disadvantage occurs to a lesser extent in shaped articles made from pellets with carbon black according to the invention.

Any type of carbon black commonly used for polypropylene can be used. Preferably a carbon black is used which has a specific surface of at least 150 m^2/g, more preferably at least 200 m^2/g (determined according to the BET method using N_2 absorption).

It is also possible for the pellets to contain a filler. The filler is preferably incorporated into the sleeve of the pellets. This will ensure that despite the presence of the filler a high production rate of the pellets is achieved. The pellets can for example contain 10-30 wt.% filler. If the pellets contain a filler, the total percentage by weight of filler and glass fibres is preferably 25-80 wt.%, more preferably 35-50 wt.%.

Examples of suitable fillers are talc, wolastonite, chalk, barium sulphate, mica, clay, etc. Preferably talc is used.

The use of a filler in the pellets according to the invention opens up additional application

possibilities.

In a further preferred embodiment the pellets according to the invention contain one or more bonding agents to improve the bond between the glass fibres and the polypropylene matrix in the shaped articles that can be made from the pellets. Examples of suitable bonding agents are propylene polymers grafted with maleic anhydride. The bonding agent preferably has a lower viscosity than the polymer composition of the fibre. The quantity of bonding agent contained in the pellets is usually between 0.2 and 5 wt.% and depends for example on the type of bonding agent and the quantity and type of glass fibres. The bonding agent is preferably incorporated into the polypropylene fibres of the core of the pellets. This will ensure that the bonding agent is highly effective.

As fibre bundle use can be made of, for example, a bundle of parallel-oriented fibres with a few twists per linear metre. The bundle is preferably made by spinning a number of glass fibres and a number of polypropylene fibres in the usual manner, distributing the fibres and then bundling them. Examples of suitable techniques for distributing the fibres are described in EP-A-616055, EP-A-59969 and EP-A-505275.

For manufacturing the pellets, the fibre bundle can be provided with a sleeve using a known cable-covering unit. A cable-covering unit generally comprises an extruder for melting the polymer composition forming the sleeve and an extruder head through which fibre bundles of the core are to be passed, the polymer composition of the sleeve being extruded around the core in the extruder head or just outside it. Next, the extrudate can be cooled by pulling the resulting strand through a water bath and the cooled strand can be reduced to the pellets, for example by chopping the strand with the aid of a

granulator.

The fibre bundles of the core are preferably passed through the extruder head of the covering unit at a speed of at least 100 m/min., more preferably at a speed of at least 200 m/min.

The strand is preferably reduced to the pellets when the temperature of the strand has been cooled to 50-100°C. The length of the pellets is preferably 5 - 50 mm, more preferably 10 - 30 mm. The strand is preferably chopped into granulate with the aid of a granulator with a rotary knife and a stator, there being no clearance between the knife and the stator. This can be achieved by, for example, spring-supporting the knife on the stator. The advantage of this is that the strand can be cut well without, for example, fibres from the core being pulled out of the sleeve.

The pellets according to the invention can be processed into shaped articles by the usual methods, e.g. pressing and injection moulding and extrusion.

The invention also relates to a process for processing the pellets according to the invention into shaped articles by

- melting the pellets in an extruder
- extruding the resulting melt into one or more objects
- placing the object or objects in the opened mould for the shaped article while still molten
- closing the mould to shape the shaped article.

These operations result in shaped articles with very good mechanical properties. The shaped articles moreover have a homogeneous composition.

A single-screw extruder is preferably used for melting the pellets. The single-screw extruder preferably has a screw dimensioned in such a way that the pellets melt mainly under the influence of heat conduction from the extruder wall. The length of the

screw is preferably 15D - 40D (D being the diameter of the screw), more preferably 20D - 30D. The flight depth in the feed zone of the screw is preferably 0.13D - 0.28D. The flight depth in the pumping zone of the screw is preferably 0.08D - 0.15D.

On the head side the extruder preferably has a valve that can be closed during the melting of the pellets, there being a buffer space between the valve and the screw tip in which a sufficient quantity of melt can be collected to extrude the object. During the melting of an amount of granulate sufficient for forming the object, the screw preferably translates in such a way that the size of the buffer space increases as a function of the quantity of molten granulate and the melt is collected in the buffer space at a pressure higher than atmospheric pressure, but preferably not higher than 30 bar, more preferably not higher than 15 bar. In this way it is achieved that the granulate melts well, the glass fibres are impregnated with polypropylene melt, but nevertheless glass fibre breakage occurs to only a small extent.

Next, the valve can be opened and the object can be extruded by causing the screw to translate in the opposite direction, after which the valve is closed once again. The cross-sectional area of the die at the extruder head is preferably at least 50% of the cross-section of the extruder barrel. More preferably, the cross-sectional area of the die equals 75-100% of the barrel cross-section. The die may have any shape. The die may for example have a round, square or rectangular shape, depending on the desired shape of the object.

The object or objects are preferably placed in the mould as it/they is/are being extruded. The mould is preferably closed as soon as possible after the extrusion of the object or objects, so that the shaped article is shaped.

It is also possible to manufacture shaped

articles from mixtures of pellets according to the invention and pellets of polypropylene which do not contain glass fibres.

The invention also relates to shaped articles made of the pellets. Examples of such shaped articles are housings of electronic and electromechanical equipment, machine parts and car parts, such as bumper beams, dashboard frames, parcel shelves, fascias and seat frames.

Example 1

A fibre bundle of glass fibres and polypropylene fibres was covered with a polypropylene sleeve with the aid of an extruder fitted with an extruder head for cable covering.

The fibre bundle was composed of five "direct rovings" of 630 Tex each, each of which contained 75 wt.% glass fibres and 25 wt.% polypropylene fibres. The fibres were distributed in such a way that they were present in the fibre bundle as single fibres and in groups of fibres with at most 50 fibres. The polypropylene used for the polypropylene fibres was a homopolymer with a melt index (230°C/2.16 kg) of 21 dg/min and a density of 904 kg/m^3.
The polypropylene of the sleeve was a homopolymer with a melt index of 45 dg/min and a density of 904 kg/m^2. The polypropylene fibres contained 2 wt.% bonding agent and the sleeve contained 1 wt.% bonding agent. This means that the granulate contained 1.2 wt.% bonding agent. As bonding agent, Polybond (TM) 3150, a maleic anhydride-modified polypropylene, supplied by Uniroyal of the UK, was used.
The extruder us was a single-screw extruder supplied by Schwabenthan (TM) of Germany and had a screw diameter of 30 mm. The cable covering head was of the tubing type and the diameter of the nozzle for the fibre bundle of the cable covering head was 2.4 mm. The melt

temperature of the polypropylene of the sleeve was 240°C. The fibre bundle was passed through the head at a speed of 100 m/min (the line speed) and covered. After covering, the glass fibre content of the pellets was 30 wt.%.

The strand thus formed was passed through a water bath and thus cooled to 50°C and chopped into a granulate with a length of 12.5 mm.

The pellets were subsequently fed to a single-screw extruder from Messrs Kannegiesser of Germany having a diameter of 80 mm, a length of 26 D, a flight depth in the feed zone of 15 mm and flight depth of 9 mm in the pumping zone. On the side of the screw tip the barrel could be closed with the aid of a hydraulic valve. The pellets were melted and collected in a buffer at the screw tip of the extruder, while the screw carried out a translation such that a pressure of 15 bar was maintained in the buffer. After about 27 seconds a buffer had been formed with a weight of about 600 g of molten granulate.

Subsequently an object was extruded in about 2.7 sec. by opening the valve and causing the screw to translate in the opposite direction so that the buffer was emptied. The object had a cylindrical shape with a diameter of 85 mm and a length of 140 mm.

Immediately after having been extruded the object was placed at the centre of the mould cavity of a mould of a square, flat plate, the longitudinal direction of the object being parallel with one of the sides of the mould cavity. Next, a plate was compressed which had a length and width of 400 mm and a thickness of about 3.2 mm. The mould had a temperature of 50°C.

Test bars were cut from the plate, perpendicular to and parallel with the direction coinciding with the longitudinal direction of the object as it had been placed in the mould cavity before the plate was pressed.

The E-modulus (Emod), stress at break (σ break) and elongation at break (ϵ break) were determined in accordance with ISO R 527-1B and the Izod impact strength in accordance with ISO 180-4A.

The results are listed in Table 1. In this table the values in perpendicular direction ($\perp$) and in parallel direction (//) and the averages of these values (av.) are given.

Comparative experiment A

A granulate containing 30 wt.% glass fibres was prepared by impregnating a bundle of glass fibres of 2400 Tex via a melt impregnation process with a polypropylene homopolymer having a viscosity of 150 Pa.s, measured at a shear rate of 1 s-1 and 240°C, chopping the resulting strand of glass fibres and polymer into a granulate with a pellet length of 12.5 mm, as described in EP-A-170245. During impregnation the line speed was 10 m/min. The pellets contained 1.2 wt.% Polybond (TM) 3150.

The pellets were melted, extruded into an object and compressed to obtain a plate as described in Example 1. Subsequently the mechanical properties were measured as in Example 1. The results are also included in Table 1.

Table 1

Mechanical properties of shaped articles from granulate according to the invention and granulate from impregnated glass fibres.

	Example I			Experiment A		
	//	⊥	av.	//	⊥	av.
E mod (MPa)	7500	3200	5350	5930	3930	4930
σ break (MPa)	136	45	91	101	65	83
ϵ break (%)	2.4	2.7	2.6	2.2	2.7	2.5
Izod ($KJ.m^2$)	62	42	53	54	34	44

Example II

Example II was carried out in the same way as Example I, except that the pellets contained 0.12 wt.% carbon black. The carbon black used was Elftex (TM) 460, supplied by Messrs. Cabot of the USA, having a specific surface of 74 m^2/g (BET, N_2 absorption). The carbon black was incorporated into the sleeve of the pellets.
The results are given in Table 2.

Comparative experiment B

Comparative experiment B was carried out in the same way as comparative experiment A, except that the pellets contained 0.12 wt.% carbon black. The carbon black used was Elftex (TM) 460.
The results are given in Table 2.
A comparison of the results from Tables 1 and 2 shows that as a result of the addition of the carbon black the mechanical properties deteriorate, but that this

deterioration is less pronounced in the pellets according to the invention.

Table 2

Mechanical properties of shaped articles from granulate according to the invention and granulate from impregnated glass fibres with carbon black.

	Example II			Experiment B		
	//	⊥	av.	//	⊥	av.
E mod (MPa)	5900	3700	4800	5400	3700	4550
σ break (MPa)	87	50	69	88	47	68
ϵ break (%)	1.8	2.2	2.0	1.7	1.8	1.8
Izod ($KJ.m^2$)	38	17	28	30	16	23

Example III

Example III was carried out in the same way as Example II, except that the carbon black used was Black Pearls (TM) 800, supplied by Messrs. Cabot of the USA, having a specific surface of 210 m^2/g.
The carbon black was incorporated into the sleeve of the pellets.
The results are listed in Table 3.
A comparison of the results of Examples II and III shows that the mechanical properties are better in the case of the carbon black with the high specific surface.

Table 3

Mechanical properties of shaped articles from pellets according to the invention with carbon black with a high specific surface.

	Example III		
	//	⊥	av.
E mod (MPa)	7200	3400	5300
σ break (MPa)	130	42	86
ϵ break (%)	2.3	2.2	2.3
Izod ($KJ.m^2$)	56	32	44

C L A I M S

1. Rod-shaped pellets containing a thermoplastic polymer and glass fibres, with the glass fibres extending in the longitudinal direction of the pellet and having the length of the pellet, characterized in that the pellets have a core and a sleeve, with the core containing a mixture of glass fibres and polypropylene fibres, with the polypropylene of the fibres being a homopolymer or a random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1-olefins with 4-10 C-atoms and dienes with 4-10 C-atoms, the polypropylene of the fibres having a melt index (230°C/2.16 kg) of 5 - 500 dg/min. and a density of at least 900 kg/m^3, with at least 50 wt.% of the sleeve consisting of a polypropylene homopolymer, random copolymer of propylene and less than 10 wt.% of one or more olefins from the group ethylene, 1-olefins with 4-10 C-atoms and dienes with 4-10 C-atoms or a block copolymer of propylene and at most 27 wt.% of one or more olefins from the group ethylene and butene and at most 8 wt.% of one or more olefins from the group 1-olefins with 3-10 C-atoms and dienes with 4-10 C-atoms and the propylene contained in the sleeve having a melt index of 1 - 200 dg/min. (230° C/2.16 kg) and a density of at least 900 kg/m^3.

2. Pellets according to claim 1, characterized in that the value of the melt index of the polypropylene of the fibres is higher than that of the polymer composition of the sleeve.

3. Pellets according to claim 1 or 2, characterized in that the glass fibres are as single fibres

and/or as fibre bundles with at most 300 fibres.

4. Pellets according to claim 3, characterized in that the glass fibres and the polypropylene fibres are present as single fibres and/or a fibre bundles with at most 100 fibres.

5. Pellets according to claim 3 or 4, characterized in that at most 50% of the glass fibres are entirely surrounded by glass fibres.

6. Pellets according to any one of the claims 1-5, characterized in that the sleeve contains 75-100 wt.% polypropylene.

7. Pellets according to any one of the claims 1-6, characterized in that the pellets contain 0.01-4.0 wt.% carbon black.

8. Pellets according to claim 7, characterized in that a carbon black type is used which has a specific surface of at least 150 m^2/g.

9. Process for th preparation of rod-shaped pellets according to any one of the claims 1-8, characterized in that a strand is chopped into the pellets with the aid of a rotary knife and a stator, there being no clearance between the knife and the stator.

10. Process for processing the pellets according to any one of the claims 1-8 into shaped articles by melting the pellets in an extruder, extruding the resulting melt into one or more objects, placing the object or the objects in the opened mould for the shaped article while still in molten condition, and closing the mould to shape the shaped article.

11. Shaped articles made from pellets according to any one of the claims 1-8.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER
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According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 B29B C08J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	EP 0 628 596 A (MITSUBISHI PETROCHEMICAL COMPANY LIMITED) 14 December 1994 see the whole document see page 7, line 25 - page 8, line 34 ---	1-6,10, 11
Y	PATENT ABSTRACTS OF JAPAN vol. 95, no. 3, 28 April 1995 & JP 06 345917 A (SHOWA DENKO K.K.), 20 December 1994, see abstract & DATABASE WPI Section Ch, Week 9510 Derwent Publications Ltd., London, GB; Class A, AN 95-070427 (10) & JP 06 345 917 A (SHOWA DENKO K.K.) , 20 December 1994 see abstract --- -/--	1-6,10, 11

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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1

Date of the actual completion of the international search	Date of mailing of the international search report
12 January 1998	16/01/1998

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Molto Pinol, F

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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